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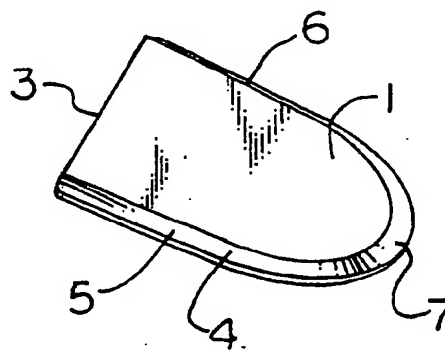
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(54) **Substrates containing a molded adhesive layer and method for preparing same**

(57) This invention provides rigid substrates containing on at least one surface a layer of an adhesive composition that is applied during an injection molding operation. The degree of bonding between the adhesive layer and the substrate is greater than can be achieved using conventional methods for coating substrates with adhesive compositions.

**Fig. 4**



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## Description

This invention relates to substrates containing a layer of adhesive on at least one surface. More particularly, this invention relates to rigid substrates wherein a layer of adhesive is cohesively bonded to at least one surface of the substrate during a molding operation. This invention also relates to a novel method for preparing these adhesive-coated substrates.

The process of coating or encapsulating organic and inorganic substrates with molten materials while the substrates are confined in a mold cavity having the contour of the final coated article is described in the patent and journal literature. These materials are non-tacky in the solid state.

The prior art of this disclosure is generally represented by the following publications: JP-A(s) 54/32565, 54/102355, 56/142045, 62/253418, 63/111267, 63/270107, 1/280517 and 5/269919; JP-B 64/16657, NL-A 85/01897 and US-A(s) 4,327,369, 4,386,042, 4,810,590, 5,002,548 and 5,160,780.

Methods taught in the prior art for bonding two substrates along a common interface typically comprise the steps of applying a layer of an adhesive in the form of a liquid or a film to at least one of the surfaces to be bonded, bringing these surfaces into contact with one another and maintaining the surfaces in close contact, preferably under pressure, during solidification and/or curing of the adhesive. The adhesive can be applied by spraying, dipping or placing a preformed layer of the adhesive on one of the surfaces to be bonded.

A shortcoming of laminates prepared by applying a preformed adhesive to one of the substrates to be bonded is the absence of strong cohesive bonding between the adhesive and the substrate to which the adhesive is initially applied. An objective of this invention is to improve the bonding between a substrate and a layer of adhesive that is applied to at least one surface of the substrate.

The objectives of the present invention are achieved by molding a layer of adhesive onto at least one surface of a rigid substrate. The surface(s) of the substrate to be coated forms at least one surface of the mold cavity into which the adhesive is injected. The contour of the mold cavity corresponds to the contour of the adhesive layer.

This invention provides a method for bonding a layer of an adhesive composition to the upper surface of a rigid substrate having upper and lower surfaces, said method comprising

- 1) forming an enclosed mold cavity from an open cavity mold and said rigid substrate wherein said upper surface mates with said open cavity mold;
- 2) injecting an adhesive composition which is non-flowing under ambient conditions into said mold cavity under a pressure and at a temperature sufficient to cause said composition to flow and completely fill said enclosed mold cavity and bond to

said upper surface;

3) converting the resultant layer of said composition to a non-flowable material; and

4) separating the substrate coated with said composition from said open cavity mold.

The adhesive compositions used in the present method contain at least one film-forming ingredient that is typically an organic polymer or a polyorganosiloxane.

The polymer is preferably curable by a crosslinking reaction and the composition contains a crosslinking agent for the polymer.

A significant feature of the present invention is the molding process which is used to apply the adhesive composition to the substrate. This process results in a substantial improvement in bond strength relative to prior art methods for applying adhesives to substrates as molten materials, as solutions or as preformed films.

Molding the adhesive on the substrate also improves bonding between the adhesive and a second substrate to which the adhesive-coated surface of the substrate is subsequently applied to form an integrally bonded laminate.

In a preferred embodiment of the present invention, the substrate to which the adhesive layer is applied during our molding operation is a metal or ceramic mounting base for a rear view mirror of a motor vehicle and the adhesive layer is used to bond the base to the windshield of the vehicle.

Substantially no restrictions apply to the type of material used to form the rigid substrate onto which a layer of adhesive composition is molded.

Preferred substrates for coating with the curable adhesive composition are metals, most preferably steel or aluminum, engineering plastics such as polyesters, polycarbonates and polysulfones and other types of organic polymers. The polymers preferably contain glass fibers or other reinforcing agents to render them substantially non-deformable under the pressures and at the temperatures required to cause the adhesive composition to flow during the molding operation.

The dimensions of the rigid substrate are likewise not critical with respect to the present method for applying a curable adhesive composition. For a practical molding operation, it is desirable that the surface to be coated is less than one meter in any dimension.

The surface on to which the adhesive is molded is preferably planar.

A preferred substrate is depicted in the accompanying drawings.

Figure 1 is a top view of a preferred substrate for coating with an adhesive layer of the present method. This substrate is described in the example of the present specification.

Figure 2 is a side view of the substrate of Figure 1 along line 2-2.

Figure 3 is a sectional view of the substrate of Figure 1 along line 3-3 and

Figure 4 is a perspective view of the substrate of

Figure 1.

The substrate depicted in the drawings is a block comprising an upper surface 1 and a lower surface 2. These surfaces are connected by a minor planar side 3 and a major side 4 consisting of two planar sections 5 and 6 joined by a curved section 7. Surfaces 1 and 2 are planar and substantially parallel. The block is formed from a rigid material such as a metal or an organic polymer with optional reinforcing agents to provide the desired rigidity.

In a preferred embodiment of the present invention, wherein the block is used as the mounting plate for an automotive rear view mirror, the curved section of side 4 exhibits a radius of curvature of from 0.2 to 1 inch (0.5 to 2.5 cm).

The dimensions and contours of the upper and lower surfaces and the sides of the block depicted in Figures 1-4 are not critical with respect to the present method and will be determined by the end-use application of the adhesively coated substrate prepared using this method.

The length and width of upper surface 1 can be from 0.5 to 39 inches (1.3 cm to 1 meter) or more and the block can be from 0.1 to 10 inch (0.25 to 25 cm) or more in thickness.

The orientation of sections 5 and 6 of side 4, with respect to one another, is not critical with respect to the ability of at least one surface of the block to be coated by the present invention. Sections 5 and 6 can be substantially parallel with respect to one another, or, as shown in the accompanying drawings, the sections can converge slightly toward one another with the result that dimension *a* is greater than dimension *b* by 0.1 mm or more.

In the preferred embodiment depicted in the accompanying drawings sides 3 and 4 are beveled at an angle of from 10 to 45 degrees, with the result that the dimensions of side 1 are somewhat smaller than the corresponding dimensions of side 2.

The adhesive compositions that are molded to form our claimed adhesive-coated substrates contain at least one film-forming ingredient that is any of the known organic or organosilicon polymers exhibiting adhesive properties. If the adhesive properties of the polymer(s) are insufficient to provide the desired bond strength, the compositions contain one or more of the known tackifying resins and/or adhesion promoters to improve the bonding strength of our composition. Polymers disclosed in the prior art as film type adhesives include, but are not limited to, natural and synthetic organic elastomers and curable organosiloxane compositions containing known adhesion-promoting ingredients.

The requirements of adhesive compositions of the present method are 1) the ability of the composition to flow sufficiently to form a coherent coating on the non-deformable substrate during a molding operation; 2) the ability of the composition to cohesively bond to the surface that the composition is in contact with during the molding operation and to any substrate with which the

adhesive layer is placed in contact before curing of the composition; and 3) that the adhesive composition not exhibit measurable flow, as measured during a 24 hour period under ambient temperature and pressure, typically 25°C. and a pressure of one atmosphere (760 mm of mercury). Curable adhesive compositions are preferably non-flowing under ambient temperature and pressure before being cured.

The temperature during the molding operation must be below the decomposition temperature of all ingredients in our adhesive composition. If the composition is curable, the temperature must be also be below the initiation temperature of the curing reaction.

Preferred adhesive compositions are solid under ambient conditions and flow without curing at temperatures from 25 to 100°C. under gauge pressures of from 100 to 2000 pounds per square inch (0.69 to 13.8 MPa). As used herein, the term "solid" refers to materials exhibiting a dry, tacky surface.

Preferred adhesive compositions of this invention are curable and comprise a curable organic polymer or polyorganosiloxane together with any additional curing agents and/or curing catalysts.

Organic polymers that exhibit sufficient adhesion, either alone or in combination with tackifying resins, such as esters of rosin acids, qualify them for use as the film-forming ingredient of our moldable adhesive compositions include, but are not limited to, thermoplastic polyolefins such as polyethylene and polypropylene; polyesters, including polycarbonates, that are prepared using condensation or free radical reactions; polyamides; polyurethanes; polysulfones; plasticized polymers of vinyl esters and vinyl acetals such as polyvinyl acetate and polyvinyl butyral; epoxide polymers; phenolics; nitrile elastomers; polyimides; and copolymers of ethylene and ethylenically unsaturated carboxylic acids such as acrylic acid.

Polyorganosiloxanes that cure to form elastomeric materials are preferred for use as the polymer ingredient(s) of the adhesive composition based on their processability and bonding properties. The adhesive properties of compositions containing heat curable film-forming organosiloxane polymers, such as polydiorganosiloxanes, is improved by the presence of coupling agents; adhesion promoters; and/or organosiloxane resins containing  $R_3SiO_{1/2}$  and  $SiO_{4/2}$  units, where R represents a monovalent hydrocarbon radical, most preferably an alkyl radical containing from 1 to 4 carbon atoms. Combinations of these organosiloxane resins with elastomeric polyorganosiloxanes constitute a known class of pressure sensitive adhesives.

A preferred class of compounds suitable for use as adhesion promoters and/or primers to increase the adhesion exhibited by both organic and organosiloxane polymer compositions are silanes and organosiloxanes containing at least one hydrolyzable group and one organofunctional group bonded to silicon atoms. The hydrolyzable group is preferably methoxy or ethoxy and the organofunctional group is preferably alkenyl, epoxy,

amino or (meth)acryloxyalkyl. Preferred alkenyl radical contain from 2 to 5 carbon atoms. The choice of organofunctional group will typically depend upon the type of substrate, i.e., organic or inorganic and the polymer(s) present in the adhesive composition.

To improve their bond strength and moisture resistance, the polyorganosiloxanes are preferably crosslinkable. Because the adhesive compositions are typically one part, i.e., all of the ingredients of the composition are packaged in a single container, when long term storage stability is desirable, the reaction used to crosslink the polymer is one that requires heating to achieve curing at a useful rate.

Preferred curing reactions for the present adhesive compositions are hydrosilation (the reaction between silicon-bonded hydrogen atoms present on a curing agent and alkenyl radicals present on a polyorganosiloxane) and free radical reactions initiated by the decomposition of organic peroxides with decomposition temperatures above 50°C.

In addition to at least one curable polyorganosiloxane and the optional adhesion promoter, the curable organosiloxane compositions preferably contain a reinforcing agent, a crosslinking agent and, optionally, a catalyst for the curing reaction. Finely divided forms of silica are preferred reinforcing agents. The silica can be of the fumed or precipitated type. A preferred type of reinforcing silica is prepared from an alkyl orthosilicate and is treated with organosilicon compounds containing silanol or hydrolyzable groups to prevent a phenomenon referred to as "creeping". The method for preparing this type of silica is more fully described in US-A 4,344,800.

In the present method, the adhesive composition is applied to at least one surface of the substrate during an injection molding operation. The surface(s) of the substrate to be coated defines a portion of the mold cavity.

Methods and equipment for injection molding compositions that are solids under ambient conditions and will flow when subjected to pressure with optional heating, are sufficiently described in the literature that a detailed description in this specification will serve no useful purpose. A complete discussion of this subject is contained in the Encyclopedia of Polymer Science and Engineering, edited by Herman Mark et al. and published by John Wiley and Sons.

In our method for coating rigid substrates with adhesive compositions, the surface to be coated serves as one portion of an enclosed mold cavity.

During the first step of the molding process the perimeter of an open cavity mold, that serves as the remaining portion of the enclosed mold cavity, is placed on or around the surface to be coated. The adhesive composition is retained within this enclosed cavity during the subsequent molding operation. The contours of the surfaces that constitute the open mold cavity are preferably planar.

The second step of the molding operation comprises injecting the adhesive composition into the enclosed mold cavity under sufficient pressure and with

heating as required to convert the composition to a flowable material. The amount of composition is sufficient to completely fill the enclosed mold cavity.

The gauge pressure used to inject the composition will depend at least in part on the viscosity of the composition at the temperature of the molding operation. This pressure can range from 100 to 2000 PSI (0.69 to 13.8 MPa) or higher, and is preferably from 1200 to 1700 PSI (8.3 to 11.7 MPa.). When heating is required, the temperature of the composition is typically from 30 to 150°C.

The third step of the present method involves converting the adhesive composition to a non-flowable material. If the composition has been heated during the molding step, the layer of composition should be allowed to cool to the extent that it solidifies.

The fourth step of the present method involves separating the coated substrate with its bonded layer of adhesive from the open cavity mold that formed the remaining portion of the enclosed cavity during the molding step. This step of the process is facilitated by coating the surfaces of the open cavity mold prior to the molding step of the present method, with a material that will not adhere to the adhesive composition and that will not transfer to the surface of the adhesive layer during the molding operation. Suitable releasing materials include solid fluorinated polymers such as polytetrafluoroethylene, polyvinylidene fluoride and fluorosilicone polymers.

Separation of the adhesive-coated substrate from the remainder of the mold cavity is further facilitated by exerting a positive pressure against the coated surface of the substrate. The pressure can be exerted by mechanical means such as a piston or ram or by a pressurized gas such as air or nitrogen.

Achieving adequate bonding between the adhesive composition and the substrate may require applying a primer or coupling agent to the surface to be coated prior to the molding operation and/or the presence of an adhesion promoter in the adhesive composition. Compounds suitable for use as primers and adhesion promoters are discussed in a preceding section of this specification.

If the adhesive composition is crosslinkable, the process conditions during the injection and molding steps are controlled to avoid significant crosslinking of the film-forming polymer(s) present in this composition. The adhesive composition is crosslinked after the surface of the substrate containing the molded adhesive layer is placed in contact with a second substrate to form the laminates that represent a preferred end use application for products of the present invention. These laminates consist of two substrates that are bonded to one another by a layer of one of the present adhesive compositions.

The following example describes a preferred substrate, adhesive composition and molding conditions. Unless otherwise specified, all parts and percentages in the examples are by weight and the reported viscosity

values were measured at 25°C.

The substrate was a substantially rectangular block formed by sintering finely divided steel particles to form the unitary structure shown in Figures 1-4 of the drawings.

The block was 0.17 inch (0.43 cm) thick and consisted of an upper surface 1 and lower surface 2 connected by a planar side 3 and side 4 consisting of two planar sections 5 and 6 and a curved section 7. Side 3 was 1 inch (2.5 cm) in length, the two planar sections of side 4 were 0.75 inch (1.9 cm) in length, and the radius of curvature of the curved section 7 was 0.43 inch (1.1 cm). Lower surface 2 measured 1.2 inches (3.0 cm) in length. The planar sections 5 and 6 of side 4 angled slightly toward one another in the direction of curved section 7, with the result that dimension  $a$  was 0.99 inch (2.5 cm) and dimension  $b$  was 0.90 inch (2.3 cm).

Sections 5, 6 and 7 of side 4 were beveled at an angle of 29 degrees. As a result of this beveling, the maximum length of upper surface 1 was 1.17 inch (3 cm) and the maximum width of this surface was 0.855 inch (2.2 cm).

Upper surface 1 was coated with a primer composition by spraying. The primer composition contained 4 percent by weight of allyltrimethoxysilane, 1 percent by weight of tetrabutyl titanate and 95 percent by weight of heptane.

When the primer composition had dried the block was placed on a horizontal surface with the coated upper surface (1) facing upward. The open mold cavity, used to form a portion of the enclosed mold cavity into which the adhesive composition was subsequently injected, had a perimeter corresponding in contour to the perimeter the upper surface (1) of the block but with the maximum length and width 0.02 inch (0.5 mm) shorter than the corresponding dimensions of the upper surface and a depth of 0.03 inch (0.76 mm). The sides of the open mold cavity originated from a planar surface that contained a passage for injection of material into the mold cavity and were substantially perpendicular with respect to this surface. The interior surfaces of the open mold cavity were coated with a layer of polytetrafluoroethylene as a release agent.

The perimeter of the open mold cavity was placed on the coated surface of the block such that the longitudinal axes of the block and the open mold cavity were collinear. Sufficient pressure was exerted against the exposed lower surface (2) of the block to form a seal between the surface of the block and the perimeter of the open mold cavity, thereby retaining all of the adhesive composition within the cavity during the subsequent injection molding operation. This pressure was maintained during injection of the adhesive composition.

The curable organosiloxane adhesive composition was prepared by blending the following ingredients to homogeneity:

72 parts of silica containing dimethylsiloxy, methyl-

vinylsiloxy and methylsiloxane groups on the surface of the particles, which were prepared from an alkyl silicate and the appropriate organosilicon compounds as treating agents using the method described in US-A 4,344,800;

12 parts of a dimethylvinylsiloxy-terminated polydimethylsiloxane gum containing 0.142 mole percent of methylvinylsiloxane units and exhibiting a Williams plasticity of 150 mm;

12 parts of a gum type dimethylvinylsiloxy-terminated dimethylsiloxane/methylvinylsiloxane copolymer containing 0.5 mole percent of methylvinylsiloxane units and exhibiting a Williams plasticity of 159 mm;

0.26 part of the monoallyl ether of glycerin;

0.03 part of methyl[tris(1,1-dimethyl-2-propynyl)silane];

0.14 part of a reaction product of hexachloroplatinic acid and sym-tetramethyldivinylidisiloxane that had been diluted with a liquid dimethylvinylsiloxy terminated polydimethylsiloxane in an amount sufficient to achieve a platinum content of 0.7 weight percent; 0.5 part of 3-methacryloxypropyltrimethoxysilane; 0.5 part of 3-glycidoxypolytrimethoxysilane; and 1.9 parts of a liquid trimethylsiloxy-terminated polymethylhydrogensiloxane with a silicon-bonded hydrogen content of 1.6 weight percent.

A quantity of this curable composition sufficient to completely fill the enclosed mold cavity was pumped into the cavity under a gauge pressure of 1600 PSI (11 MPa). A jet of air under a gauge pressure of 75 PSI (0.5 MPa) was then admitted to the cavity to separate the coated block from the remaining portion of the enclosed mold cavity.

A second open mold cavity, with the same dimensions as the one used to apply the coating, was placed on the coated surface of the block in the same manner as the first cavity and sufficient pressure was applied against the layer of adhesive to flatten the sprue remaining from the molding operation into the body of the adhesive composition. A stream of air under a gauge pressure of 75 PSI (0.5 MPa) was then directed into the enclosed mold cavity to eject the coated block.

After the blocks were stored for 24 hours, test samples were prepared by pressing the coated surface of a block against a sheet of plate glass using a force of 100 pounds (45 kg). The glass sheet containing the adhered block was then placed in an oven maintained at a temperature of 141°C. for one hour to cure the adhesive layer.

After cooling to ambient temperature, one group of five samples (A) was tested for adhesion by securing a clamp around the beveled edge of the block, securing the glass sheet in position and exerting sufficient tensile force on the clamp to pull the block away from the glass plate. The force required to break this bond was recorded.

A second group of five samples (B) were placed in

room temperature water for 24 hours before being tested.

For comparative purposes, the same curable adhesive composition that had been molded onto the upper surface 1 of the blocks was prepared in the form of a calendared sheet measuring 0.03 inch (0.76 mm) in thickness. Pieces, corresponding in contour to the upper surface (1) of the block, were cut from the sheet and applied to this surface together with an overlying release layer using manual pressure or a hand-held roller. The blocks used for the comparative example were the same ones described in the preceding section of this example and depicted in Figures 1 to 4 of the accompanying drawings.

After the layer of release material was removed, the adhesive-coated side of the block was then placed against a sheet of glass, the adhesive was cured and adhesion of the block to the glass was evaluated as described above.

The results of the adhesion test are recorded in the table. The comparative samples, tested prior to and following water immersion, are referred to as C and D, respectively.

Sample No.	Force in Pounds (Kg)
A	451.4 (204.9)
B	310.3 (140.9)
C	411.0 (186.6)
D	197.8 (89.8)

The greater force required to separate blocks A and B, prepared using the present method, from the glass relative to comparative samples C and D clearly demonstrate the improved bonding is achieved by molding our adhesive coating onto the substrate rather than applying a pre-cut sheet of adhesive to the block as taught in the prior art.

#### Claims

1. A method for bonding a layer of an adhesive composition to a rigid substrate having an upper surface, said method comprising:

- 1) forming an enclosed mold cavity from an open cavity mold and said rigid substrate wherein said upper surface mates with said open cavity mold to form said enclosed cavity;
- 2) injecting an adhesive composition which is non-flowing under ambient conditions into said enclosed mold cavity under a pressure and at a temperature sufficient to cause said composition to flow and completely fill said enclosed mold cavity and bond to said upper surface;

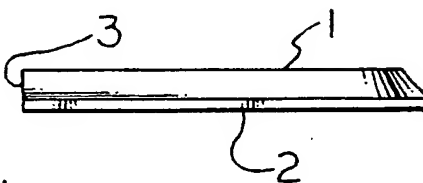
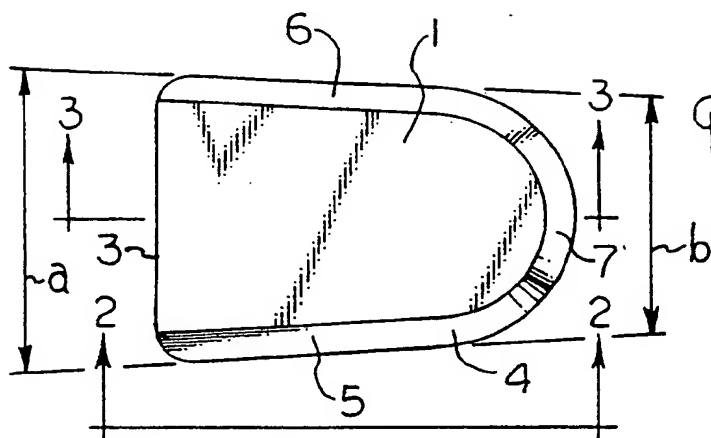
- 3) converting the resultant layer of said composition to a non-flowable material; and

- 4) separating the substrate coated with said composition from said open cavity mold.

2. A method according to claim 1 wherein said rigid substrate is formed from at least one metal or at least one organic polymer; said adhesive composition comprises a film-forming polymer selected from organic polymers or polyorganosiloxanes; said composition is injected into said enclosed mold cavity under a gauge pressure of from 100 to 2000 PSI (0.69 - 13.8 MPa) and at a temperature of from ambient to 200°C.; and the substrate coated with said composition is separated from said open cavity mold by exerting a positive pressure against the coated upper surface of said substrate.
3. A method according to claim 2 wherein said rigid substrate is formed from at least one metal; the surfaces other than said substrate that constitute said mold cavity are planar and contain a coating of a non-transferable release agent; said composition is injected into said mold cavity under a gauge pressure of from 1200 to 1700 PSI (8.3 to 11.7 MPa) and at a temperature of from 30 to 150°C.; and said positive pressure is exerted by a pressurized gas.
4. A method according to claim 3 wherein said adhesive composition cures by a platinum catalyzed hydrosilation reaction and contains at least one adhesion-promoting additive; said adhesion-promoting additive and said primer composition comprise a silane or a liquid polyorganosiloxane containing at least one hydrolyzable group bonded to a silicon atom; and said release agent is a fluorocarbon polymer or a fluorosilicone polymer.
5. A coated substrate obtainable by the methods of claims 1 to 4.

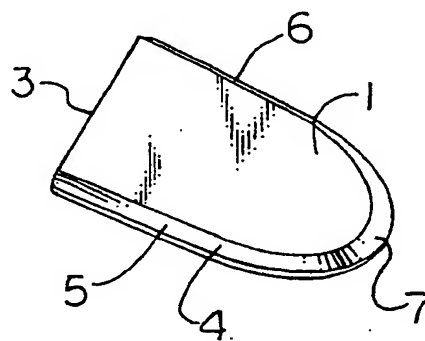
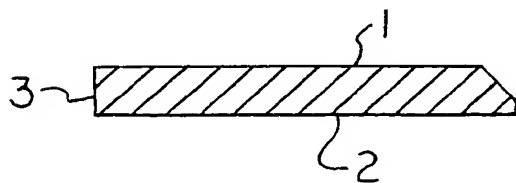
*Fig. 1*

*Fig. 2*



*Fig. 3*

*Fig. 4*





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## EUROPEAN SEARCH REPORT

Application Number  
EP 96 11 4918

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
X	WO-A-95 21734 (TETRA LAVAL HOLDINGS & FINANCE ) 17 August 1995	1,5	B29C45/14 C09J183/04
Y	* the whole document *	4	
A	---	3	
X	PATENT ABSTRACTS OF JAPAN vol. 13, no. 153 (M-813) [3501] , 13 April 1989 & JP-A-63 312818 (TOAGOSEI CHEM IND CO LTD), 21 December 1988, * abstract *	1,5	
P,X P,A	DE-A-44 46 027 (HENKEL KGAA) 4 July 1996 * the whole document *	1,2,5 3	<div>TECHNICAL FIELDS SEARCHED (Int.Cl.6)</div> <div>B29C C09J</div>
Y	EP-A-0 639 622 (DOW CORNING CORP.) 22 February 1995 * the whole document *	4	
A	DE-A-19 60 764 (KÜBA KÜHLERFABRIK BAUERBRUNN) 9 June 1971 * the whole document *	1,5	
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The present search report has been drawn up for all claims			
Place of search <b>THE HAGUE</b>		Date of completion of the search <b>10 January 1997</b>	Examiner <b>Bollen, J</b>
<b>CATEGORY OF CITED DOCUMENTS</b> X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		I : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	

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